Applicability of Fermi golden rule and possibility of low-field runaway transport in nitrides

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In order to justify applicability of the standard approach of perturbation theory for the description of transport phenomena in wide-band polar semiconductors with strong electron-phonon interactions, we have compared dependences of energy losses to the lattice on the electron drift velocity obtained for different materials in the frameworks of (a) a perturbative approach based on calculation of the scattering rates from Fermi's golden rule and (b) a non-perturbative approach based on the path-integral formalism of Thornber and Feynman. Our results reveal that despite strong electron-phonon coupling in GaN and AlN such that intercollision times become of the order of the period of phonon oscillation, standard perturbative treatment can still be applied successfully for this type of material. Our findings also indicate possibility for unique long-distance runaway transport in nitrides which may occur at the pre-threshold electric fields. Polaron ground state energy and effective masses are calculated for GaN and AlN as well as for GaAs and Al_2O_3.

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I. INTRODUCTION

One of the main factors determining electron transport characteristics in polar semiconductors is scattering of the electrons by polar optical phonons. For relatively weak electron-phonon interactions, when scattering events can be considered as independent, use of Fermi's golden rule for the calculation of energy-dependent frequencies of electron transitions provides an adequate description of experimentally obtained velocity-field curves. Upon increasing of the interaction, however, the polaronic effects induced by autolocalization of an electron by the inertial part of the crystal polarization become more prominent and they determine the character of the scattering. Intensification of the electron-phonon interaction leads eventually to a situation where the average intercollision time becomes less than the duration of a collision. Such a strong coupling, therefore, requires proper account for the quantum interference effects and makes the problem of electron drift essentially nonlinear. This complicates dramatically the theoretical treatment of carrier scattering and field-dependent transport since, for the given case, the standard perturbation technique is not applicable.

To ensure energy conservation for the short-time perturbations, the inverse scattering rate, τ, must be large enough to satisfy the inequality $\tau \gg \hbar/\Delta\mathcal{E}$, where $\Delta\mathcal{E}$ is the electron transition energy. This criterion, however, does not allow one to conclude whether or not a wide class of materials in which the transition energy may become of the order of the linewidth can be described successfully in the framework of standard perturbation approaches. To such a class of materials belong, in particular, nitrides of Al and Ga. These semiconductors have been investigated recently quite intensively due to a number of their unique properties that can be utilized in the current state-of-the-art semiconductor technology. However, most attempts to describe scattering processes in nitrides have been undertaken assuming the validity of Fermi golden rule. Moreover, despite the possibility of growing the nitrides of group III in zincblende-like structures, their crystal structure at ambient conditions is wurtzite-like. For wurtzites, it is generally necessary to account for optical anisotropy when considering the carrier-optical-phonon interactions. Since optical phonon spectra in wurtzites are far more complicated than those of cubic crystals, the majority of theoretical results have been obtained by ignoring the features of the phonons in optically anisotropic media. Recently, a formalism has been developed[1] for evaluation of the scattering rates in bulk wurtzite-like semiconductors and heterostructures by taking into account peculiarities of the phonon spectra obtained in the framework of macroscopic dielectric continuum model.[2] Due to the anisotropy-induced complexity of the problem, Ref. 1 took advantage of the perturbation theory. However, as indicated by the previously-discussed considerations, the validity of such an approach requires independent confirmation.

The present paper demonstrates applicability of Fermi's golden rule for describing adequately the electron-longitudinal-phonon interaction in polar materials by comparing the field-velocity dependences obtained in the frameworks of (a) the perturbation theory and (b) the non-perturbative path-integral approach of Thornber and Feynman[3] (TF). A supplemental, but very important, result obtained from the present investigation is the discovery of the possibility for unique low-field long-distance runaway transport in materials characterized by

strong electron-phonon interactions. Examples of such materials are the nitrides of Al and Ga.

II. MODEL

The most systematic and self-consistent approach for evaluating the long-range polaron ground-state energy G, effective mass m_0, and carrier energy dissipation for both strong- and weak-coupling limits of electron-phonon interaction has been developed by Feynman et al.[3,4,5] In Ref. 3, the problem of electron drift in a parabolic band under steady state conditions is considered quantum mechanically[6] assuming that all the energy losses are due to interaction of electrons with polar optical modes. Taking advantage of the Fröhlich's polaron model, the authors used the path-integral method to eliminate the lattice coordinates from the momentum balance equation and obtained an expression for the magnitude of the electric field E that is required to maintain a particular magnitude of electron velocity V at arbitrary temperature and interaction strength characterized by the coupling constant

$$\alpha = \frac{e^2}{\hbar}\left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0}\right)\left[\frac{m^*}{2\hbar\Omega}\right]^{1/2}. \qquad (1)$$

Herein, e is the elementary charge. Evaluation of α requires four parameters: electron effective mass m^*; frequency of the longitudinal phonon Ω; static dielectric constant ϵ_0; and high frequency dielectric constant ϵ_∞. All these parameters can be measured experimentally and they are the only external parameters required for calculation of energy loss per unit distance eE versus V.

Thornber and Feynman[3] have calculated the dependences of $eE(V)$ for three coupling constants ($\alpha = 3, 5, 7$) over a wide range of reciprocal temperatures $\beta = \hbar\Omega/(k_B T)$, where k_B is the Boltzmann constant and T is the temperature of the lattice. The general result of these calculations can be summarized briefly as follows. For each particular α, $eE(V)$ has a maximum at some threshold value V_{th}. For $\beta > 1$, location of this maximum becomes independent of temperature. For $V < V_{th}$, eE is an increasing function of velocity. This interval of velocities corresponds to a stable situation when energy loss to the lattice due to the absorption and emission of optical phonons can be compensated by the energy gained by the electron from the applied field in such a way that at the given E, a small deviation ΔV of the velocity from its steady state value V_s creates a force, $e[E(V_s) - E(V_s \pm \Delta V)]$, which stabilizes the velocity at V_s. When the external field approaches the value $E_{th} = E(V_{th})$, the dependence tends to saturate since the magnitude of the energy loss due to interaction with optical phonons is finite. The case $E > E_{th}$ was excluded from consideration because no steady state conditions can be reached for such fields and electron would accelerate infinitely. The theory, however, predicts the existence of solutions for $V > V_{th}$. In this region, eE is a decreasing function of the velocity which leads to an unstable steady state situation. For this case, any deviation of the velocity from V_s would lead to either deceleration of the electron to velocity $V < V_{th}$ which is stable at the given field, or a *gradually* increasing acceleration if ΔV leads to an increase in velocity. It is essential, that for $V > V_{th}$, the dependence $eE(V)$ can be interpreted as a time-dependent momentum loss in the absence of the external field. This loss would coincide with the rate of electron momentum loss if the criterion $dV/dt \ll V/(\text{duration of the collision})$ is satisfied. For $\beta > 1$, $eE(V > V_{th})$ also becomes independent on temperature.

In order to simplify the comparison, and taking into account that the strongest electron-polar-optical-phonon scattering is due to emission of the longitudinal optical (LO) phonons, we will consider the case when $\beta \approx 4$. Due to high energy of LO phonons in the nitrides, such value of β would correspond to room temperature in these materials. For GaAs, which we take as a reference point in our investigation, $\beta = 4$ would correspond to lattice temperature of order of 104 K. Since β_{GaAs} is slightly higher than 1 at the room temperature, the result obtained for maximum energy loss per unit distance can be compared to the experimental *velocity-field* dependences (see, for example, Ref. 7 and citations therein). Indeed, at some threshold field E_{th}, the dependence $V(E)$ has a maximum caused by transitions of the carriers to an upper valley with a higher effective mass. In terms of the TF model, these transitions would start to occur when the energy supply from the external field would exceed the maximum loss to the lattice; i.e., at $E > E_{th}$.

Thus, if the average kinetic energy of electrons obtained in the framework of TF model at E_{th} does not exceed the energy of bands separation and the effects of the increased effective mass due to non-parabolicity of Γ band can be neglected, the value of the argument at maximum of $V(E)$ dependence has to correlate with the extremum of the $eE(V)$.

Under the assumptions made above, we use the simplest model for estimation of energy loss to the lattice in the framework of the perturbation theory. In this model, we calculate the dependence of scattering rate $1/\tau$ due to the emission of LO phonons on a single electron kinetic energy $\mathcal{E}$ using the Fermi's golden rule. For an optically isotropic material, the scattering rate is

$$\frac{1}{\tau} = \left(\frac{2m^*}{\mathcal{E}}\right)^{1/2} \frac{e^2\Omega(N_q+1)}{\hbar}\left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0}\right) \ln\left[\sqrt{\frac{\mathcal{E}}{\hbar\Omega} - 1} + \sqrt{\frac{\mathcal{E}}{\hbar\Omega}}\right], \qquad (2)$$

where N_q is the phonon occupation number. Then, we assume that a carrier with velocity $V = \sqrt{2\mathcal{E}/m^*}$ loses the energy $\hbar\Omega$ to the lattice in a distance τV.

III. RESULTS AND DISCUSSION

A. Limiting cases

The energy loss per unit distance vs. the electron velocity calculated for GaAs at room temperature in the framework of the model which uses Fermi's golden rule is shown on Fig. 1 by the thin solid line. It is important, that as anticipated, the maximum of the dependence is in a good agreement with the experimental data which give the maximum of $V(E)$ at $E \approx 3.4-3.9$ kV/cm. One should note, that since calculations are made in a one-electron approximation and for single parabolic band, they overestimate the velocity obtained at the maximum. The experimentally measured values at maximum of $V(E)$[7] reflect the averaging of the velocities over bands with different effective masses as well as effects of non-parabolicity.

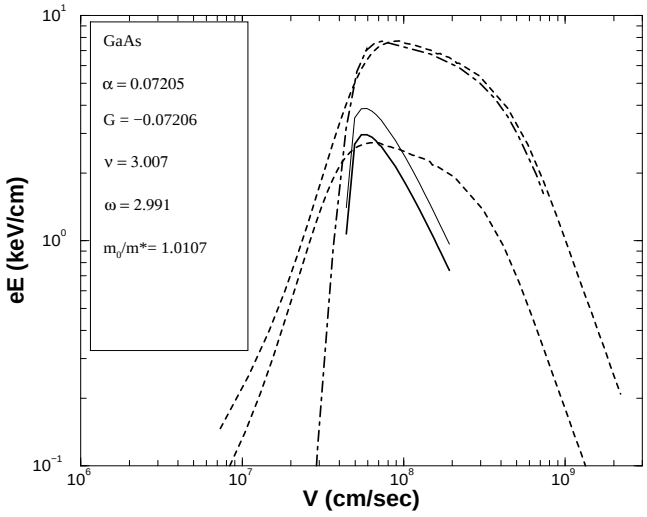

FIG. 1. The energy loss per unit distance vs. electron velocity in GaAs. Perturbative model: room temperature - thin solid line; T=104 K - thick solid line. TF model: T=104 K - upper dashed line; T ≈ 20 K - dash-dotted line. Corrected TF model: T=104 K - lower dashed line. Polaron parameters are given in the inset.

The curve calculated in the simplest model at 104 K ($\beta = 4$) is given by the thick solid line. Surprisingly, the $eE(V)$ dependence computed for $\beta = 4$ in the framework of the TF model (upper dashed line) exhibits a maximum located at somewhat higher fields. We assume that such a discrepancy occurs because in a weak-coupling limit the zero-order distribution for the electrons in this model reduces to a drifted quasi-Maxwellian. An essential requirement for such a distribution to be valid in the given case is the presence of high electron concentration and strong electron-electron interactions, which provide randomization of the direction of electron momentum between the scattering events.[8] It is unlikely, however, that such a randomization can be achieved for scattering with emission of polar optical phonons. Indeed, in the weak-coupling limit the motion of the carrier in the near-threshold fields becomes essentially one-dimensional. Conservation of energy and momenta, valid in the weak-coupling limit, require the angle between the carrier and scattered phonon momenta to be no more than $\arccos(\hbar\Omega/\mathcal{E})$. Due to this condition, emission of an LO phonon at threshold fields causes deviation of the direction of electron momentum from the direction of applied field by no more than ≈ 20 degrees. Additional focusing of the electron momentum in the direction of the electric field comes from the inverse dependence of the interaction matrix element on the phonon wavevector[9] and leads to overestimation of energy losses when using the Maxwellian distribution.[10]

In order to resolve this discrepancy, one can suggest - in analogy to the classical case - that at the same electron temperature, reduction of the dimension would correspond to a reduction in the average carrier energy. Since the deviation of the direction of the electron momentum relative to the direction of the electric field is small but finite, for the weak-coupling limit we have reduced the energy scale in TF model by a factor of two in order to match the maxima. This corresponds to decreasing the velocity and energy losses by factors $2^{1/2}$ and $2^{3/2}$, respectively. The corrected curve $eE(V)$ is shown by lower dashed line.

The differences in the shapes of the curves obtained in the simplest model and the TF model occur for the following reasons. The former model considers only the emission mechanism, whereas the latter model also takes into account absorption. As shown in the figure by dash-dotted line calculated for the uncorrected case of TF model for $\beta \approx 21$, elimination of phonons absorption by reducing the lattice temperature yields the same slope of $eE(V < V_{th})$ as in the simple model. As expected, for this temperature interval, the temperature decrease does not affect the shape of the curve at and beyond the maximum. For $V > V_{th}$, the discrepancy in $eE(V)$ between the models appears to be due to the relatively high value of dV/dt in the unstable region. In the framework of the TF model, one can estimate this value by eE/m_0, where m_0 can be obtained as $m_0 \approx m^*\nu^2/\omega^2$; ν and ω are the parameters of TF model. We have computed these parameters from minimization of the free energy at zero temperature.[4] Our estimations show that assuming the duration of the collision to be equal to τ, the value of the derivative would be much less than V/τ only for $V \sim 10^9$ cm/sec. Thus, for $V > V_{th}$ the simple model cannot be used for GaAs under the conditions when an external field is applied.

Additional confirmation of the idea that the corrections required for the application of the TF model in the

weak-coupling limit are induced by focusing of the carrier momentum comes from the fact, that in the case of strong coupling - when the directions of momenta are randomized due to strong electron-phonon interaction discussed previously - the model correctly explains the experimentally-obtained results. In Fig. 2 we depict the energy losses calculated for Al_2O_3. The maximum on the dependence obtained in the TF model (dashed curve) is in the excellent agreement with the experimentally obtained maximum losses in this material, 0.03 eV/Å.[11]

$$\alpha(\theta) = \frac{e^2}{\epsilon^{\infty}\Omega}\sqrt{\frac{m^*}{2(\hbar\Omega)^3}}\left[\frac{\omega_{Lz}^2-\omega_z^2}{(\Omega^2-\omega_z^2)^2}\cos^2\theta + \frac{\omega_{Lt}^2-\omega_t^2}{(\Omega^2-\omega_t^2)^2}\sin^2\theta\right]^{-1}, \qquad (3)$$

where ω_{Lz}, ω_z, ω_{Lt}, and ω_t are the characteristic frequencies of the A1(LO), A1(TO), E1(LO), and E1(TO) modes, respectively. The phonon frequency as a function of θ can be obtained from the dispersion relation for the extraordinary bulk phonons.[2] The dependence $\alpha(\theta)$ calculated for GaN is shown on Fig. 3.

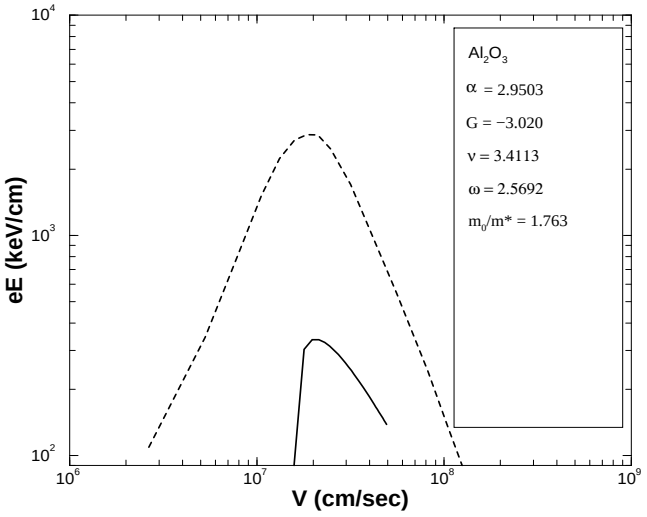

FIG. 2. The energy loss per unit distance vs. electron velocity in Al_2O_3 at room temperature. Perturbative model: solid line; TF model: dashed line; Polaron parameters are given in the inset.

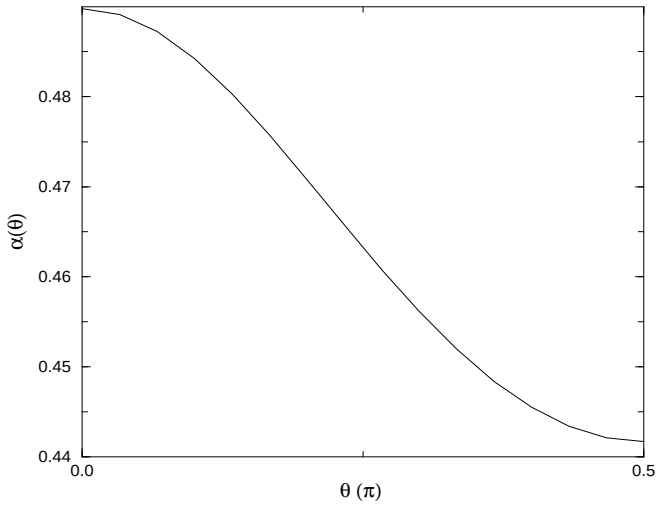

FIG. 3. Dependence of the polaron coupling parameter on the angle between the phonon wavevector and the optical axis in GaN.

As expected, the maximum losses calculated in the perturbative model (solid line) are less by an order of magnitude. Of course, since for the given material $\alpha > 1$, the simple model is not valid and we have presented here both dependences simply as a means of estimating the possible error which can be induced by a perturbative treatment and to demonstrate that no energy scale reduction can fit these dependences.

B. Intermediate case: nitrides

Due to the optical anisotropy inherent to wurtzites, the coupling parameter of polaron theory α becomes dependent on the angle θ between phonon wavevector and the optical axis. Assuming that $\epsilon_z^{\infty} = \epsilon_t^{\infty}$, we define this dependence as

To obtain the $eE(V)$ dependence in the TF model we have used $\alpha = 0.46$. This value corresponds to the energy of LO phonon calculated for GaN in the cubic phase.[12] The energy of the LO phonon in cubic AlN is taken to be 113 meV which is - as for the GaN case - between the energies of A1 and E1 LO modes in wurtzite phase.[2] The scattering rates are calculated according to the formalism developed in Ref. 1. Comparison of the dependences obtained for the nitrides is given in Fig. 4. Again, the maxima obtained in the simple model correlate very well with the threshold electric fields of $V(E)$ dependences computed in the Monte Carlo technique[13] for a three-valley model for the conduction band: 140 kV/cm for GaN and 450 kV/cm for AlN. Note that in order to match the maxima one needs to use the same reduction of the energy scale when calculating the energy losses in the TF model as for the case of GaAs. Additionally, one can see that the shapes of the dependences for $V \geq V_{th}$ are almost the same. This agreement between the simple and

the TF models is due to the extremely short duration of the collisions which can be estimated roughly as the inverted scattering rate, $\tau \sim 10^{-14}$ sec. The increase of the polaron effective mass cannot compensate the increase in the energy loss near the threshold value and, therefore, cannot reduce dV/dt. Nevertheless, due to frequent collisions, the criterion $dV/dt \ll V/\tau$ is satisfied for the nitrides even at $V \to V_{th}^{+}$.

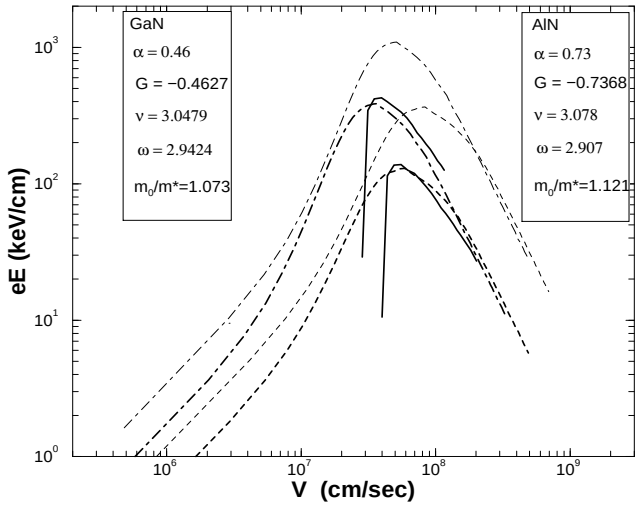

FIG. 4. The energy loss per unit distance vs. electron velocity in GaN and AlN calculated at the room temperature. Perturbative model: AlN - upper solid line, GaN - lower solid line; TF model: AlN - dash-dotted lines, GaN - dashed lines. Thin and thick broken curves are the uncorrected and corrected dependencies, respectively. Polaron parameters are given in the insets.

The comparison made here allowed us to conclude that in materials for which $\tau\Omega \gtrsim 1$, application of Fermi golden rule to explain transport phenomena is as good as in materials traditionally handled with the perturbation theory, i.e., in the materials for which much stronger criterion, $\tau\Omega \gg 1$, is satisfied.

C. Low-field runaway transport

It is important to emphasize another result of our investigation. Figure 5 represents the dependencies of energy losses on electron kinetic energy obtained in the framework of the TF model at room temperature for the three materials considered: GaAs, GaN and AlN. On the figure, vertical arrows indicate the energy of the closest upper valley in the corresponding conduction band. The picture shows clearly the possibility to achieve unique pre - threshold - field runaway transport in the nitrides. Indeed, let us consider a GaN sample in an external electric field of 100 kV/cm. As shown in the figure, the two steady-state solutions for electron energy would correspond to such a situation. One of the solutions lies in the region $V < V_{th}$ and, consequently, it reflects a stable solution with respect to the electron energy fluctuations. Another one falls into the unstable area, $V > V_{th}$. Suppose an electron is injected into the sample with an energy somewhere in between the energy which corresponds to the second, unstable, solution and the threshold energy.

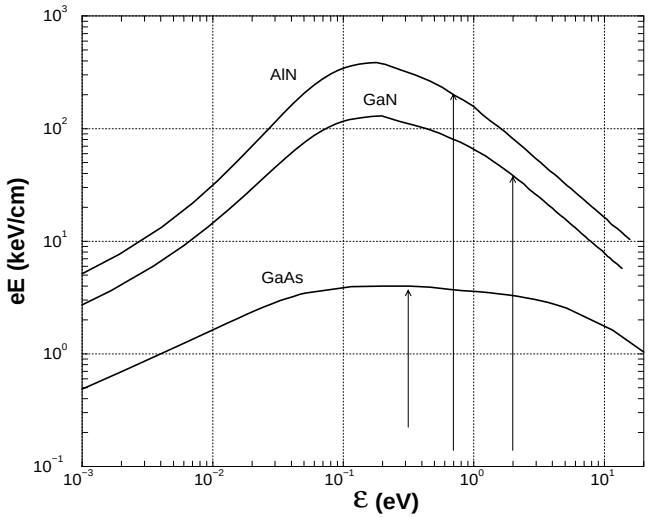

FIG. 5. The energy loss per unit distance vs. electron kinetic energy. Vertical arrows indicate the energy of the closest upper valley in the corresponding conduction band. T = 300 K.

In this case, since the energy losses to the lattice would exceed the energy gain from the external field, the electron would decelerate until the stable solution at given field would be reached. If, however, the energy of the injected electron would just slightly exceed the value of high-energy steady-state solution, the electron would accelerate moving downwards on the unstable branch of the dependence until it gains enough energy, $\mathcal{E}^*$, to appear in the nearest upper valley of the conduction band. The value of acceleration would gradually increase due to the increasing difference between the force caused by the external field, $F = 100$ keV/cm, and the force caused by energy losses to the lattice represented by the $eE(\mathcal{E})$ dependence. In order to estimate the minimum runaway length L_r , we have assumed that the energy of the injected electron is 0.7 eV and that the maximum force, $F - eE(\mathcal{E}^*)$, is constantly applied to the carrier. Under this assumption, we obtained $L_r^{GaN} > 220$ nm.

It is easy to see that because the intervalley separa-

tion in AlN is smaller, the effect in this material is expected to be not as strong as in the previous case. Assuming injection energy 0.31 eV and applying field 300 kV/cm, we get $L_r^{AlN} > 39$ nm. Our results also show that the previously-discussed runaway transport cannot be achieved in GaAs due to the small intervalley gap and broad peak on the $eE(\mathcal{E})$ dependence.

The results presented in this paper were obtained ignoring the non-parabolicity effects. These effects, however, would not change our main findings qualitatively. In order to improve accuracy of the expected quantities, further investigation is required. It is interesting to note, that the abstract possibility of the low-field runaway transport was mentioned initially by Thornber and Feynman.[3] Nitrides of Ga and Al promise to be materials where such transport could be actually realized.

IV. SUMMARY

In present paper, we have compared the energy losses to the lattice calculated in different polar semiconductors within the frameworks of both non-perturbative and perturbative approaches. Our results reveal that application of Fermi golden rule for calculation of the scattering rates in nitrides, where $\tau\Omega \approx 1.5$, is as appropriate as application of this standard perturbative treatment in the materials for which the well-known criterion $\tau\Omega \gg 1$ is satisfied. This finding dramatically simplifies the analysis of transport phenomena in the wide-band polar semiconductors with intermediate magnitude of polaron coupling factor, $\alpha < 1$.

Applying the non-perturbative path-integral approach of Thornber and Feynman to evaluation of field dependent electron energy dissipation in AlN, GaN and GaAs, we have found that pre-threshold low-field runaway electron transport can be realized in the nitrides. The conditions for such a transport can be formulated as follows: (a) the energy of the injected carrier should exceed the energy which corresponds to the solution of the momentum balance equation located on the unstable branch of $eE(V)$; and (b) the separation between the energy of injected carrier and the energy of the bottom of an upper valley must be high enough to provide a finite value of runaway length. It must be at least a few times higher than the energy of the polar optical phonon.

V. ACKNOWLEDGMENTS

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